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Biodiesel production from waste frying oils: Optimization of reaction parameters and determination of fuel properties

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ABSTRACT

Alkali-catalyzed transesterification of waste frying oils (WFO) was carried out in various conditions to investigate the effects of catalyst concentration, reaction time, methanol/oil molar ratio, reaction temperature, catalyst type (hydroxides, methoxides and ethoxides), and purification type (such as washing with hot water, purification with silica gel and dowex) on the biodiesel yields. The optimum conditions were 0.5% wt. of NaOH, 30 min reaction time, 50 °C reaction temperature, 7.5 methanol to oil ratio and purification with hot distilled water. 96% biodiesel yield with ~97% ester content was obtained within in these conditions, and the activation energy was found to be as 11741 J mol^{-1}. The determined specifications of obtained biodiesel according to ASTM D 6751 and EN 14214 standards were in accordance with the required limits. As a conclusion, the present study indicates that WFO derived fuel promises being an alternative for petrodiesel, and could be used in engines without a major modification due to its qualifications.

1. Introduction

Biodiesel production has been receiving global attention since petroleum prices are increasing continuously. Moreover, biodiesel production might be a solution for preventing petroleum dependency and environmental concerns. As biodiesel is gaining considerable demand, standards are vital for its commercialization and market introduction. The commercialization of biodiesel in many countries around the world has been accompanied by the development of standards to ensure high product quality and user confidence. Fuel grade biodiesel must be produced to meet most referred industrial specifications, which are the European Standard Specifications for Biodiesel (EN 14214) and the American Standard Specifications for Biodiesel Fuel (B100) Blend Stock for Distillate Fuels (ASTM 6751) [1,2].

Biodiesel can be produced from various vegetable oils and animal fats. However, the raw material costs and limited availability of vegetable oil feed stocks are problems for the biodiesel production. The high cost of vegetable oils, which could be up to 75% of the manufacturing cost, has led to the production costs of biodiesel becoming approximately 1.5 times higher than that of petrodiesel. The growing population of developing countries like Turkey has increased the food consumption and therewith the production of a large amount of waste frying oils (WFO)/fats. Due to high amount of oil consumption, Turkey has approximately 3.5×10^5–4.5×10^5 tones/year waste frying oil (WFO) capacity and this amount is increasing every year [3,4]. Note that the utilization of all waste oils as domestic animal feed is banned and this is enforced by EU, during frying process, many harmful compounds are formed [5]. The conversion of this huge amount of WFO into the fuel eliminates the environmental impacts caused by the harmful disposal of these waste oils, such as into drains. Moreover, it can significantly reduce the total manufacturing cost, because the price of WFO is 2–3 times cheaper than virgin vegetable oils [1,6–8]. As known, optimization of production conditions is very important in a large-scale production process in order to maximize the yield of biodiesel and minimize the production costs. Therefore in this extensive study, the conversion of WFO into biodiesel is optimized with regarding to process parameters and purification types. The fuel properties are also investigated according to the accepted standard methods and compared with ASTM D 6751 and EN 14214 biodiesel standards.

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2. Experimental

2.1. Materials

WFO samples, as feedstock, were collected from local restaurants and then filtered to remove any inorganic residues and suspended matters. All the chemicals used in experiments were purchased from Merck chemicals. The certified reference standards for Gas Chromatography (GC) were purchased from Sigma-Aldrich.

2.2. Analytical methods

Properties of WFO were determined according to the AOCS standard test methods of oil and fat analysis [9]. Fatty acid composition of WFO, methyl ester, mono-di-triglyceride and methanol content of produced biodiesel were determined using Hewlett-Packard 7890 Series II Gas Chromatography. The fuel characteristics of the final biodiesel product were determined according to the ASTM D 6751, EN 14214 and AOCS standard test methods.

2.3. Transesterification procedure

The system used for transesterification reactions consisted of temperature bath, reaction flask with reflux condenser and rpm controlled mechanical stirrer. Experiments were carried out in a 500 cm^3 two necked batch reactor, one for condenser and the other for samples. A digital temperature bath (Julabo ED-5) with a PID controller was capable of maintaining the reaction temperature to within ±0.03 °C. The used oil was preheated until the selected temperature was reached. After that, a mixture of methanol and catalyst was added to the oil and the transesterification reaction began. The stirrer was set at 600 rpm to avoid the mass transfer limitations and a magnetic bar was used for stirring. The reaction was timed as soon as the methanol/catalyst mixture was added and continued for 30 min. At the end of the reaction, the product was kept overnight for cool down and separation of phases. The upper phase consisted of methyl esters, and the lower phase contained the glycerol, the remaining catalyst, the excess methanol, the soaps formed and some entrained methyl esters. Fewer concentrations of catalyst, glycerol and methanol were in the upper methyl ester phase. After removing the glycerol from the methyl ester phase using the separation funnel, the upper methyl ester phase was washed with hot distilled water, at 50 °C, with a ratio of 1:1, for two times without stirring and three times with stirring [6]. The washed methyl esters were than dried over sodium sulfate and filtered under vacuum and then weighed to find the biodiesel yields.

All the experiments were carried out at least three times in order to determine the variability of the results and to assess the experimental errors. In this way, the arithmetical averages were calculated for all the results which had an experimental error of less than ±0.5%. Experiments was repeated at various conditions such as, catalyst concentration (0.4–1.5 wt.% of WFO), reaction time (5–35 min), methanol/oil molar ratio (3:1–12:1), reaction temperature (25–75 °C), catalyst type (hydroxides, methoxides and ethoxides), and purification type (refining steps such as washing with hot water, purification with silica gel and dowex) were studied.

3. Results and discussion

3.1. Properties of waste frying oil

The physical and chemical properties of the WFO were analyzed and given in Table 1. As it can be seen from Table 1, the oil contents mostly oleic and linoleic acids and the properties of oil are in the range of standard limits. Therefore, the WFO is efficient for biodiesel production.

3.2. Effects of catalyst concentration on biodiesel yield

The catalyst concentration is an essential parameter for process optimization to make the process feasible. In this part, the reaction temperature of 70 °C, reaction time of 30 min, and methanol/oil molar ratio of 6:1, and agitation intensity of 600 rpm were maintained as constant with varying catalyst (NaOH) concentrations (0.4–1.5 wt.% of WFO) for the production of WFO methyl ester. The results are given in Fig. 1(a). As shown, increasing the concentration of catalyst produces lower biodiesel yields. According to the experimental results, the maximum biodiesel yield was obtained as 93.8% with 0.5 wt.% NaOH. This can be explained as addition of excess amount of catalyst caused more triglycerides participating in the saponification reaction and caused the formation of soap in presence of high catalyst concentration and a slight decrease in biodiesel yields. This result was in accordance with the previous works found in literature [7–12].

3.3. Effects of reaction time on biodiesel yield

In this part, to determine the effect of reaction time on formation of methyl esters, experiments were carried out at various reaction times ranging from 5 to 35 min at catalyst concentration of 0.5 wt.% NaOH, reaction temperature of 70 °C, molar ratio of methanol to oil 6:1, and agitation intensity of 600 rpm. The experimental results are shown in Fig. 1(b). As seen, biodiesel yield increased with reaction time at the beginning, reached at a yield of 93.0% in the first 15 min and a maximum yield at 30 min as 93.9%, and then decreased slightly with increasing reaction time. Because longer reaction times led the hydrolysis of esters and caused more fatty acids to produce soap [10]. According to our results, reaction time had a significant effect on the conversion of the triglycerides up to 30 min, but increasing further the reaction time had decreased the biodiesel yield. Due to reaching the maximum biodiesel yield as 93.9%, 30 min was selected as the optimal reaction time for WFO transesterification in the present conditions.

3.4. Effects of methanol/oil molar ratio on biodiesel yield

In order to investigate the effect of molar ratio on biodiesel yields, experiments were conducted with various molar ratios ranging from 3:1 to 12:1 with a constant catalyst concentration of 0.5 wt.% NaOH, reaction time of 30 min, reaction temperature of 70 °C, and agitation intensity of 600 rpm. Experimental results are

Table 1
The physical and chemical properties of the WFO.

Property	Value	Limits (AOCS)
Density (kg/m^3, 15 °C)	963.51	942
Refractive index (20 °C)	1.473	1.422
Peroxide number (mg peroxide/g)	1.350	2.160
Acid index (mg KOH/g)	0.587	1.24
Iodine index (g I^-/100 g)	122	118–141
Saponification index	185	[a]
Viscosity (cp)	63.5	[a]
Fatty acid composition	wt.%	
Palmitic acid (C16:0)	7.07	
Stearic acid (C18:0)	2.42	
Oleic acid (C18:1)	36.68	
Linoleic acid (C18:2)	52.20	
Docasanoic acid (C22:0)	0.83	

[a] Not specified.

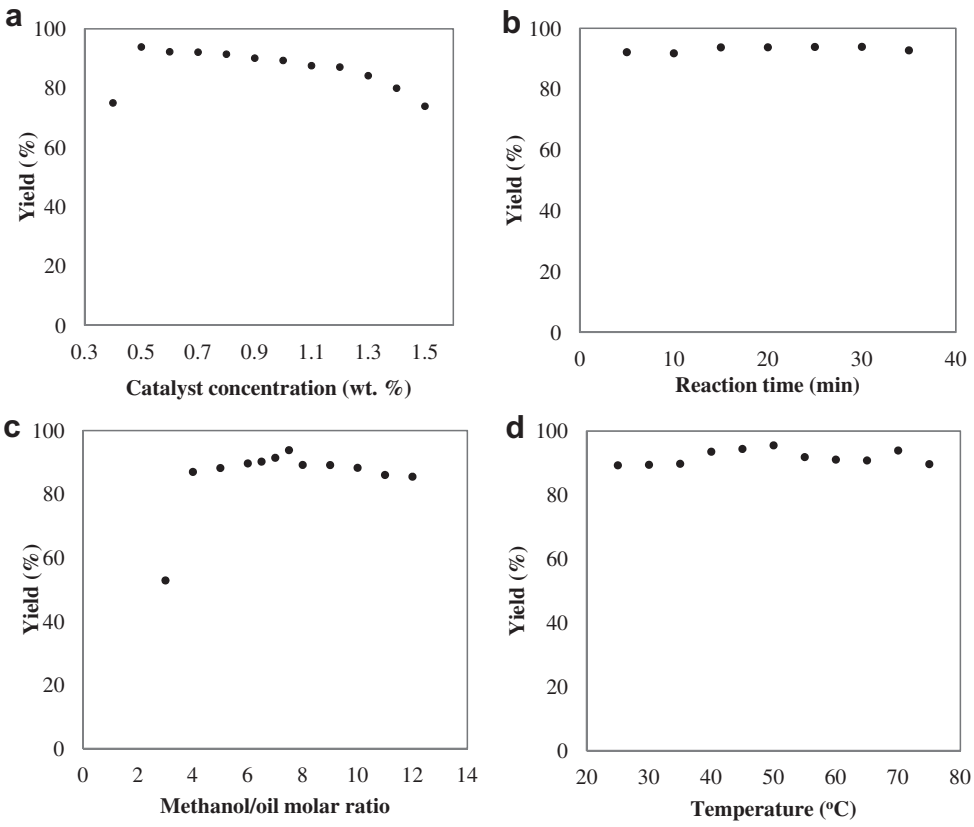

Fig. 1. Biodiesel yields: effects of catalyst concentration (a), reaction time (b), methanol/oil molar ratio (c), and reaction temperature (d).

presented in Fig. 1(c). As seen, the stoichiometric ratio (3:1) gave the lowest biodiesel yield as 52.9%. During the experimental study, the maximum biodiesel yield was determined between 6:1 and 8:1 methanol:oil molar ratio. Previous studies reported that the optimum methanol:oil molar ratio should be as minimum as 6:1 [13,14]. As seen from Fig. 1(c) the maximum biodiesel yield was obtained at a methanol:oil molar ratio of 7.5:1 as 93.4%, so that this ratio could be selected as optimum molar ratio for further part of the study. Nevertheless, increasing the molar ratio more than 8:1 reduced the biodiesel yields. Since excess methanol increased the solubility of glycerol, this caused the interference of a little amount of glycerol with the ester phase; consequently the diluted glycerol in ester phase leaded more foam formation with the lower product yields [15].

3.5. Effects of reaction temperature on biodiesel yield

In this part, to determine the effect of reaction temperature on biodiesel yields, experiments were carried out at various temperatures ranging from 25 to 75 °C in the presence of 0.5 wt.% NaOH catalyst at a reaction time of 30 min, methanol/oil molar ratio of 7.5:1, and agitation intensity of 600 rpm. The results for reaction temperature optimization are represented in Fig. 1(d). Some of the previous studies reported that transesterification could proceed satisfactorily at ambient temperatures with the use of alkaline catalyst, while given enough time [6,16]. As seen from Fig. 1(d), biodiesel yield was obtained at ambient temperature as 89.30% and increased to 95.52% with increasing temperature up to 50 °C. Therefore, the optimum reaction temperature was chosen as 50 °C. After that point, there was a slight reduction in the biodiesel yields. The reaction temperature near and above boiling point of alcohol tends to accelerate the saponification of glycerides by the alkaline catalyst before completion of the alcoholysis, which is an undesirable result. Experimental results are agreed with the previous works [7,17].

As known, temperature is very effective on rate of reaction. For the very large majority of chemical reactions, increasing the temperature increases the rate of the reaction. The transesterification reaction is a pseudo-first order reaction in excess of methanol. The average overall reaction rate constant can be calculated according to the experimental data given in Fig. 2. The overall rate conversion has a relation with yield (α) and temperature as follows:

$$k = -\ln(1-\alpha)/t \quad \ln k = -E_a/RT + C$$

where E_a is the activation energy, R is the gas constant (J mol^{-1} K^{-1}), T is the absolute temperature, and C is a constant [18]. Fig. 2 gives

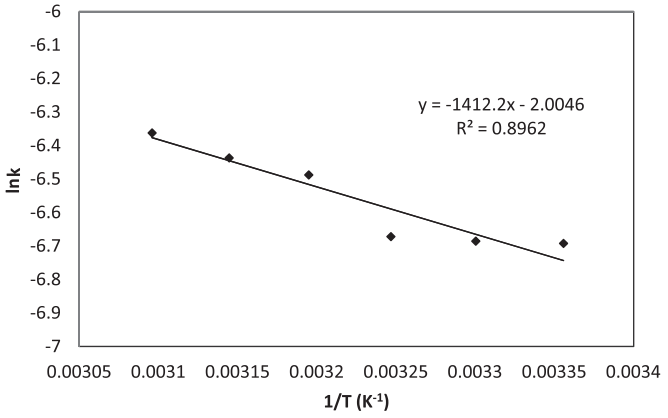

Fig. 2. Plot of ln k vs $1/T$ for the transesterification reaction.

the plot of $\ln k$ versus $1/T$ for transesterification reaction. Linear regression analysis of these data gives a slope of −1412 with a correlation coefficient of 0.896. From the plot of $\ln k$ versus $1/T$, the slope is equal to $(-E_a/R)$. Thus, a value for E_a of 11,741 J mol^{-1} was calculated for the reaction.

3.6. Effects of catalyst type on biodiesel yield

The reaction can be catalyzed by alkalis, acids or enzymes. Due to the low cost, higher reaction rates, moderated conditions, NaOH and KOH are the most commonly used alkali catalysts. Nonetheless, the alkali homogeneous catalysts are highly hygroscopic and absorb moisture from the air. Besides, their use in transesterification of oils produces soap by neutralizing the free fatty acid in the oil or triglyceride saponification. Both soap formations are undesirable side-reactions, because they partially consume catalyst, which decrease the yield and make the separation and purification steps more difficult [17]. Therefore in this study, a comparison is made among the various basic catalysts to investigate the effects of alkali type on the biodiesel yields. NaOH was compared with KOH, CH_3ONa, CH_3OK, C_2H_5ONa, and C_2H_5OK and results are given in Fig. 3. As seen from the figure, sodium derived catalysts give higher yields than potassium derived ones. This can be explained by the fact that sodium has a lower molecular weight which leads higher transesterification reaction rates [18]. Although the maximum biodiesel yield was obtained as ~97% in presence of sodium ethoxide catalyst, NaOH was selected as the most efficient catalyst since it is the cheapest and gives high yields. This result is in agreement with several works indicating that the alkoxides of alkaline metals are more effective than the corresponding hydroxides, although for economic reasons sodium hydroxide is more often used [7,17].

3.7. Effect of purification type on biodiesel yield

The refining step of the products obtained by the transesterification is very important because the purity level of the biodiesel has strong influence on its fuel properties. Especially, the amount of glycerides and triglycerides present in the fuel can cause serious problems in application. According to the European Union standards for alternative diesel fuel, the contents of free fatty acids, methanol, glycerol, and water in the biodiesel are restricted and biodiesel must be at least 96.5% pure [15]. Up to this part, all samples were purified with washing hot distilled water at 50 °C. The other purification methods are also investigated by treating the mixture with: (a) silica gel, (b) 5 wt.% phosphoric acid, (c) Dowex. There is a few study on purification methods in literature [15,19].

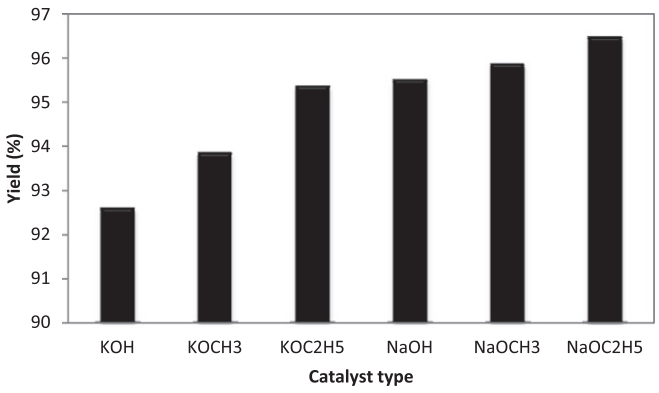

Fig. 3. Effect of catalyst type on biodiesel yield.

Table 2
The fuel properties of the used WFO methyl ester.

Property	Value	EN 14214	ASTM D 6751
Density (kg/m^3, 15 °C)	890	860–900	[a]
Refractive index (20 °C)	1.446	[a]	[a]
Acid index (mg KOH/g)	0.271	<0.50	<0.80
Saponification index	175.16	[a]	[a]
Kinematic viscosity (mm^2/s)	4.36	3.5–5.0	1.9–6.0
Cetane number	53.4	>51	>47
Flash point (°C)	175.4	>101	>130
Pour point (°C)	−3	<0	<0
Oxidation stability (h, 110 °C)	0.91	>6.0	>3.0
Cold filter plugging point (°C)	−5	<+5 (summer) >−15 (winter)	[a]
Water content (mg/kg)	480	<500	<500
Copper strip corrosion (3 h, 50 °C)	1	<1	<3
HHV (MJ/kg)	45.34	[a]	[a]
Ester content (%m/m)	96.8	>96.5	[a]
Monoglyceride content (% m/m)	0.50	<0.80	[a]
Diglyceride content (% m/m)	0.20	<0.20	[a]
Triglyceride content (% m/m)	<0.10	<0.20	[a]
Free glycerol (%m/m)	0.01	<0.02	<0.02
Total glycerol (%m/m)	0.17	<0.25	<0.24
Methanol content (%m/m)	0.44	<0.20	<0.20
Linoleic acid methyl ester (%m/m)	0.10	<12.0	[a]
Polyunsaturated methyl esters (≥4 double bound) (%m/m)	0	<1.0	[a]
Fatty acid composition	wt.%		
Myristic acid (C14:0)	0.10		
Palmitic acid (C16:0)	6.39		
Palmitoleic acid (C16:1)	0.20		
Stearic acid (C18:0)	3.57		
Oleic acid (C18:1)	33.89		
Linoleic acid (C18:2)	54.86		
Ecosanoic acid (C20:1)	0.23		
Docasanoic acid (C22:0)	0.59		

[a] Not specified.

Predojevic investigated the comparison of different purification steps for biodiesel from WFO. It was determined that the average purity level with regarding the purification treatments were found as similar for silica gel (97.41%), phosphoric acid (97.16%) and hot distilled water (97.92%) [15]. In our study, purification methods except washing with distilled water caused significant reduction in biodiesel yields. Although purification with silica gel and Dowex caused high purity, they caused a decrease in biodiesel yield to 89%. This can be explained by the fact that some liquid molecules attached on solid surfaces may cause a decrease of yields. Moreover,

Table 3
Fuel properties of biodiesels from the literature obtained from virgin and waste oils.

Fuel properties							
Cetane number	Flash point (°C)	Pour point (°C)	Kinematic viscosity (mm^2/s)	HHV (Mj/kg)	Acid index (mg KOH/g)	Density (kg/m^3)	Reference
46	[a]	[a]	4.59	[a]	0.23	892	[6]
58.6	[a]	[a]	5.01	[a]	0.80	887	[6]
[a]	120	0	4.89	[a]	0.43	880	[7]
54.5	[a]	[a]	[a]	[a]	0.48	890	[8]
53	192	[a]	9.48	36.7[b]	[a]	895.3	[20]
63.9	166	[a]	6.23	42.3[b]	[a]	883.9	[20]
56.9	38	−6	4.83	39.55	0.10	888.8	[21]
[a]	160–170	[a]	4.1–6.3	[a]	0.15–0.25	864–900	[16]
54.5	171	[a]	4.23	[a]	[a]	890	[22]
[a]	[a]	[a]	5.16	39.26	[a]	887	[23]
[a]	[a]	[a]	4.92	39.48	[a]	878	[23]
52.91	176	−3	5.29–6.46	[a]	0.289	884.4	[24]
[a]	109	0	[a]	39.31	[a]	873.7	[25]
53.4	175.4	−3	4.36	45.34	0.271	890	This study

[a] Not specified.
[b] Lower heating value.

phosphoric acid treatment neutralized and stopped the reaction but again the biodiesel yields decreased slightly up to 88%. Hence, purification with hot distilled water is more suitable for obtaining high biodiesel yields among the others. In literature, Karaosmanoglu et al. also found similar result that distilled water at 50 °C was the best refining method among these methods they used (1) the dissolving with petroleum ether and then washing with hot water, (2) neutralizing with sulfuric acid [19].

3.8. Fuel properties of produced biodiesel

Analytical methods were applied on the final biodiesel product to determine the fuel characteristics and results were compared with the diesel standards of EN 14214 and ASTM D 6751. As seen from Table 2, the properties of produced biodiesel are accordance with ASTM D 6751 and EN 14214 standards. Fuel characteristics of obtained biodiesel were also compared with other reported studies from the literature given in Table 3 and it is seen that the fuel properties of the present study are better than many reported studies such as having lower viscosity, oxidation stability, pour point and higher flash point, heating value etc.

4. Conclusions

In this study, base catalyzed transesterification of WFO were studied to optimize the reaction conditions. Optimization studies could give an idea about process conditions for pilot scale applications so that wasting time and money is avoided. The reaction parameters such as catalyst concentration, reaction time, methanol/oil molar ratio, reaction temperature, catalyst type, and purification type were optimized as catalyst concentration of 0.5% wt., reaction time of 30 min, reaction temperature of 50 °C in the presence of NaOH catalyst purifying with hot distilled water. Moreover, activation energy is calculated as 11,741 J mol^{-1} in these conditions and detailed fuel properties, compositions and oxidation stability are also determined and compared with previous works in the literature. The properties of produced biodiesel are accordance with ASTM D 6751 and EN 14214 standards. According to the experimental studies it can be concluded that WFO could be a potential alternative, economically sustainable and environment friendly fuel to other common biodiesels and petrodiesel and could be used in engines without a major modification.

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References

[1] Balat M. Potential alternatives to edible oils for biodiesel production – a review of current work. Energ Convers Manage 2011;52:1479.
[2] Abdullah AZ, Salamatinia B, Mootabadi H, Bhatia S. Current status and policies on biodiesel industry in Malaysia as the world's leading producer of palm oil. Energ Policy 2009;37:5441.
[3] Rashid U, Anwar F, Moser BR, Ashraf S. Production of sunflower oil methyl esters by optimized alkali-catalyzed methanolysis. Biomass Bioenerg 2008;32:1202–5.
[4] Tuik, 2009. <http://www.tuik.gov.tr/PreHaberBultenleri.do?id=6180> (Last accessed: 05 May 2010).
[5] Lam MK, Lee KT, Mohamed AR. Homogeneous, heterogeneous and enzymatic catalysis for transesterification of high free fatty acid oil to biodiesel: a review. Biotechnol Adv 2010;28:500–18.
[6] Encinar JM, Gonzales JF, Rodriguez-reinares A. Ethanolysis of used frying oil: biodiesel preparation and characterization. Fuel Process Technol 2007;88:513–22.
[7] Phan AN, Phan TM. Biodiesel production from waste cooking oils. Fuel 2008;87:3490–6.
[8] Bhatti HN, Hanif MA, Qasim M. Ata-ur-Rehman. Biodiesel production from waste tallow. Fuel 2008;87:2961–6.
[9] Official methods and recommended practices of the American Oil Chemists Society. Champaign, IL: AOCS Press; 1998.
[10] Leung DYC, Guo Y. Transesterification of neat and used frying oil: optimization for biodiesel production. Fuel Process Technol 2006;87:883–90.
[11] Leung DYC, Wu X, Leung MKH. A review on biodiesel production using catalyzed transesterification. Appl Energ 2010;87:1083–95.
[12] Encinar JM, Gonzales JF, Rodryguez-Reinares A. Biodiesel from used frying oil: variables affecting the yields and characteristics of the biodiesel. Ind Eng Chem Res 2005;44:5491–9.
[13] Ma F, Hanna MA. Biodiesel production: a review. Bioresour Technol 1999;70:1–15.
[14] Meher LC, Sagar DV, Naik SN. Technical aspects of biodiesel production by transesterification – a review. Renew Sustain Energ Rev 2006;10:248–68.
[15] Predojevic ZJ. The production of biodiesel from waste frying oils: a comparison of different purification steps. Fuel 2008;87:3522–8.
[16] Dorado MP, Ballesteros E, Lopez FJ, Mittelbach M. Optimization of alkali-catalyzed transesterification of brassica carinata oil for biodiesel production. Energ Fuel 2004;18:77–83.
[17] Lang X, Dalai AK, Bakhshi NN, Reaney MJ, Hertz PB. Preparation and characterization of bio-diesels from various bio-oils. Bioresour Technol 2001;80:53–62.
[18] Liu X, Piao X, Wang Y, Zhu S. Calcium ethoxide as a solid base catalyst for the transesterification of soybean oil to biodiesel. Energ Fuel 2008;22:1313–7.
[19] Karaosmanoğlu F, Cığızoğlu KB, Tüter M, Ertekin S. Investigation of refining step of biodiesel production. Energ Fuel 1996;10:890–5.
[20] Vicente G, Martinez M, Aracil J. Integrated biodiesel production: a comparison of different homogeneous catalysts systems. Bioresour Technol 2004;92:297–305.
[21] Çanakçı M, Özsezen AN. Evaluating waste cooking oils as alternative diesel fuel. G.U. J Sci 2005;18:81–91.
[22] Felizardo P, Correia JN, Raposo I, Mendes JF, Berkemeier R, Bordado JM. Production of biodiesel from waste frying oils. Waste Manage 2006;26:487–94.
[23] Enweremadu CC, Mbarawa MM. Technical aspects of production and analysis of biodiesel from used cooking oil. Renew Sustain Energ Rev 2009;13:2205–24.
[24] Çetinkaya M, Karaosmanoğlu F. Optimization of base-catalyzed transesterification reaction of used cooking oil. Energ Fuel 2004;18:1888–95.
[25] Al-Widyan MI, Al-Shyoukh AO. Experimental evaluation of the transesterification of waste palm oil into biodiesel. Bioresour Technol 2002;85:253–6.